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FLUIDISED BED POLYMERISATION

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The invention relates to a process for the polymerisation of one or more monomers in a fluidised bed reactor, which reactor comprises a reaction zone which is confined at the underside by a gas distribution plate and at the top side by a virtual end surface, in which a fluidised bed is maintained between the underside and the top side and in which at least part of the gaseous stream withdrawn from the top of the reactor is cooled to a point where the stream partially condenses into a liquid, and in which at least part of the resulting two-phase stream is recycled to the reactor via an inlet which terminates in the reactor below the gas distribution plate.

Gas-phase fluidised bed polymerisation of one or more monomers, like an olefin or olefins, is effected in a usually vertical elongated reactor in which a bed of polymer particles is maintained in fluidised condition with the aid of an ascending gas stream which contains at least the gaseous monomer(s) to be polymerised. The gas stream is passed through a gas distribution plate which separates the lower part of the reactor from the reaction zone proper. In this plate there are provided perforations that suitably distribute the gas stream supplied over the reaction zone. A peripheral section of the gas distribution plate may be sealed so as to obtain a particular pressure drop at a lower flow rate of the gas. In order to prevent polymer particles from building up on such peripheral section, the seal is preferably designed as an inclined wall which extends from the gas

distribution plate to the wall of the reactor. The angle of the inclined wall to the gas distribution plate must be greater than the angle of natural repose of the polymer particles in the reactor and,

5 furthermore, is generally at least 30°, preferably at least 40° and more preferably is between 45° and 85°.

The ascending gas stream may optionally contain one or more inert gases and for example hydrogen as a chain length regulator. An important
10 objective of the addition of inert gases is to control the dew point of the gas mixture. Suitable inert gases are for example inert hydrocarbons such as (iso)butane, (iso)pentane and (iso)hexane, but also nitrogen. Such an inert gas may be added to the gas stream as a gas
15 or, in condensed form, as a liquid.

The gas stream is discharged through the top of the reactor and, after certain processing operations, fresh monomer is added to it to make up for the monomer(s) consumed in the polymerisation, and then
20 the gasstream is again supplied to the reactor as (a portion of) the ascending gas stream in order to maintain the bed.

A catalyst is also added to the bed. During the process, under the influence of the catalyst
25 present, fresh polymer is continuously formed and at the same time polymer that has formed is withdrawn from the bed, with the bed volume and mass being kept substantially constant.

The polymerisation is an exothermic
30 reaction. Heat needs to be removed continuously so as to keep the temperature in the reactor at the desired level. Such removal is effected via the gas stream

which leaves the reactor at a higher temperature than that at which it is supplied to the reactor. The superficial gas velocity in the reactor cannot be chosen to be arbitrarily large and so no arbitrarily large amount of heat can be removed. The minimum velocity is dictated by the requirement for the bed to remain fluidised. On the other hand, the velocity must not be so large that a significant amount of polymer particles are blown out through the top of the reactor. The aforementioned limits are heavily dependent on the dimensions and the density of the polymer particles present in the bed and can be determined by experiment. Practical values for the superficial gas velocity are between 0.05 and 1.0 m/sec. These requirements are elements which limit the maximum flow rate of the gas stream at the given reactor dimensions and, thus, the maximum attainable heat removal. The maximum allowable amount of heat of reaction produced, and hence the maximum amount of polymer to be produced, are limited likewise.

The detailed design and operation of fluidised bed reactors for the polymerisation of one or more olefin monomers and suitable process conditions are known per se and are described in detail in for example US-A-4,543,399 and in WO-A-94/28032.

From that same US-A-4,543,399 it is known to replenish the gas stream discharged from the reactor with fresh monomer(s) and to cool it to a point where the stream partly condenses (the so-called "condensed mode"). The two-phase stream so obtained, which because of the latent heat of evaporation of the liquid phase has a substantially larger heat removal capacity, and

so a corresponding cooling capacity, than a stream consisting solely of a gas, is recycled to the bottom of the reactor. The dew point of the two-phase stream must be lower than the temperature in the reaction zone so that the liquid can evaporate in it. In this way, the production capacity of a fluidised bed reactor appears to be substantially higher than that of reactors which use a recycle gas without condensed liquid, said reactor having otherwise equal dimensions. In the known process the maximum amount of liquid in the two-phase stream is 20 wt %. The highest figure quoted in the examples is 11.5 wt %.

From WO-A-94/28032 it is known to separate the liquid from the two-phase stream obtained on cooling of the gas stream to be recycled and to feed said liquid to the reactor separately from the gas stream. The liquid is preferably injected or atomised at a certain height into the fluidised bed proper, optionally with the aid of a gaseous propellant. In this way, according to this publication, it is possible to feed a larger amount of liquid in proportion to the amount of gas being fed. This allows an even larger amount of heat to be removed, so allowing higher polymer production with proportionally higher heat production. WO-A-94/28032 quotes a figure of 1.21 as the maximum permissible ratio of the mass of liquid feed to the mass of the total gas feed, which figure was derived from a simulated experiment.

The present invention relates to a process for the polymerisation of one or more monomers in a specific fluidised bed reactor, which reactor, at given dimensions, allows a higher liquid mass to gas mass

ratio in the feed to the reactor than in a reactor according to the prior art, both in cases where the reactors are operated under "condensed mode conditions".

5 This object is achieved by a process in which the reaction zone of the reactor is divided into two or more compartments by one or more substantially vertical partition walls extending from a point located above the gas distribution plate to a point located
10 below the end surface.

 It has been found that when in such a reactor a fluidised bed is maintained that extends, both at the top and bottom, beyond the partition walls, so that the partition walls are submerged in the
15 fluidised bed, more liquid can be supplied in proportion to the total gas feed than in the absence of a partition wall. This increases the heat removal capacity of the process, so allowing higher heat production and hence higher polymer production rates at
20 equal reactor dimensions. Even at a constant liquid to gas mass ratio in the feed to the reactor, the process of the present invention results in a higher productivity of the reactor.

 In a reactor according to the prior art the
25 ratio of the height of the fluidised bed to the diameter of the radial cross section (H/D-ratio) usually is 3 to 5 at the most. At higher ratios it has proved impossible to maintain a stable fluidised bed if, besides gas, liquid is fed to the reactor.

30 An additional advantage of a reactor having at least one partition wall is that it is now possible to choose a higher H/D-ratio for the reactor, for

instance, an H/D-ratio of greater than 5, and even up to 20, which is much higher than in the case of the known reactors, while yet maintaining a stable fluidised bed, resulting in a more controlled
5 polymerisation process. This advantage presents major engineering advantages for polymerisation reactors because they are pressure vessels.

A particularly suitable partition wall in the reactor of the invention is a pipe or hollow
10 section placed in vertical position, preferably concentric with the reactor. Since the pipe or hollow section is completely submerged in the fluidised bed, no appreciable pressure differences occur across the wall of the pipe so that the pipe may be of light-duty
15 construction. This applies also to walls of different shapes.

The walls can simply be suspended from a higher section of the reactor, supported by a bottom section or secured to the wall of the reactor. In the
20 present context a hollow section differs from a pipe in terms of the shape of its cross section. The cross section of a pipe is curved, for example circular or elliptical, whilst that of a hollow section is angular, for example triangular, rectangular, octagonal or with
25 more angles, with or without the angles being uniformly divided. The hollow section or the pipe may have a uniform and/or tapered cross-section, for instance a cone shape, including tapering inwardly and outwardly, for instance, in a hyperbolic shape. For conical
30 shapes, it is preferred that the apex angle formed by the walls of pipe or hollow section is generally not more than 5°, preferably not more than 2.5°.

Particularly suitable are angles between 0° and 2° . The ratio of the area of the radial cross section of the pipe or hollow section to that of the reactor is between 1:9 and 9:10 and, in order to achieve as high a stability as possible, preferably between 1:5 and 3:4. In the case of a conical pipe or hollow section, the same applies to the average cross-sectional area thereof. The lower end of the pipe or hollow section is located at least $0.1 \times$ the diameter of the reaction zone above the gas distribution plate and preferably $3 \times$ that diameter at the most. If the dimensions given here are departed from, the favourable effect of the presence of a vertical partition wall is diminished. The upper end is located at least $0.1 \times$ the diameter of the reaction zone below the end of that reaction zone and preferably not more than $3 \times$ that diameter. It has been found that it is far less critical for the bed to extend further beyond the partition wall at the upper end than at the lower end. The upper end of the partition wall may be lower accordingly as the H/D-ratio of the fluid bed increases. What is stated here on the positioning of the wall in the reaction zone applies also to the vertical partition walls to be explained below.

Another embodiment of a suitable partition wall is a substantially axially oriented flat, curved or folded plate present in the reaction zone. It is preferred for such a partition wall to connect to the inner wall of the reactor although a clearance of up to 10 cm in-between is permissible. In this way, the reaction zone is divided into two or more compartments, which may be differently sized. The area ratio of the

radial cross section of a compartment to the radial cross section of the reactor preferably is between 0.1 and 0.9 and more preferably between 0.20 and 0.75. The substantially axially oriented wall should be virtually
5 vertical. This should be understood to mean parallel with the axis of the reactor in its vertical position but also out of parallel by not more than 5°, preferably not more than 2.5°.

The aforementioned beneficial effects of a
10 partition wall occur when there is a common inlet for a gas/liquid mixture at the underside of the reactor, as described in US-A-4,543,399, and also when there is a separate gas and liquid inlet in the fluidised bed, as described in WO-A-94/28032.

15 In the latter case, the liquid may be supplied to the fluidised bed via the underside of the reactor at one or more points through the gas distribution plate as well as at one or more points through the side wall. It is in any case advantageous
20 to arrange the means of introduction of the liquid in such a way that the bulk of the liquid can be supplied into the fluidised bed in a zone located under or in the central compartment if a pipe or hollow section is employed, or under or into one of the compartments if
25 one or more vertical partition walls are present. In the case of introduction of the liquid via one or more points through the side wall of the reactor, and if the partition wall is a pipe or hollow section, it is advantageous to position the means of introduction so
30 that the liquid can be supplied to the fluidised bed at a point below the lower end of the pipe or hollow section. In that case, for example by suitably choosing

the feed velocity, the liquid can be supplied to both the central compartment and the peripheral compartment of the reactor. Preferably, the bulk of the liquid is supplied to the central compartment, located within the
5 pipe or hollow section, inasmuch as the best results are obtained herewith.

In the case that a vertical plate is used as a partition wall, the liquid may be introduced in the aforementioned manner from a height below the lower
10 end of the wall but also via inlets arranged at different heights in the section of the reactor wall which confines the compartment or compartments to which the liquid is to be supplied.

The liquid is preferably injected in finely
15 divided form, preferably in atomised form, optionally with the aid of a propellant, for which purpose for example recycle gas or fresh monomer gas may be used. Injection should take place in such a way that the liquid enters the desired compartment whence it is
20 taken up by an ascending fluidising gas stream. This has been found to be favourable in terms of the amount of liquid that can be supplied to the fluidised bed without sintering of polymer particles or other undesired disturbances occurring in the bed.

25 Introducing the recycle liquid via several inlets at different heights of the reactor gives the possibility to vary the concentration of the different ingredients of the liquid inlet (through the addition of more or less monomer make up, etc.) which improves
30 the operating window of the polymerisation reaction and therefore broadening the product capabilities of the fluidised bed reactor.

In processes in which the H/D-ratio of the reactor exceeds 5, the means of introducing the liquid may also be positioned above the upper end of the partition wall(s) provided that the distance between
5 the said upper end and the top of the fluidised bed is not less than approx. 2 m.

The process of the present invention has been found to allow the benefits related to the new reactor design to fully manifest themselves. In the
10 process of the invention the reactor can be operated in a stable manner even when the mass ratio of (liquid supplied to the reactor) : (amount of gas supplied to the reactor) is higher than 2:1 or even higher than 4:1. The aforementioned ratio is in any case at least
15 10% and even more than 50% to even more than 100% higher than when the process is operated in a similar reactor without partition wall(s).

The supplied amount of gas includes, besides the gas supplied via the recycle stream, all
20 other gases supplied to the reactor, including at least the propellant and carrier gases that are employed in introducing the catalyst, a catalyst activator and/or other substances desired or needed for the polymerisation and those used for atomising the
25 supplied liquid.

The process according to the present invention is suitable for any kind of exothermic polymerization reaction in the gas phase. Suitable monomer include olefin monomers, polar vinyl monomers,
30 diene monomers and acetylene monomers. The process of the present invention is especially suitable for the manufacture of polyolefins by the polymerisation of one

or more olefin monomers, at least one of which is preferably ethylene or propylene. Preferred olefin monomers for use in the process of the present invention are those having from 2 to 8 carbon atoms.

5 However, small quantities of olefin monomers having more than 8 carbon atoms, for example 9 to 18 carbon atoms, can be employed if desired. Thus, in a preferred mode, it is possible to produce homopolymers of ethylene and/or propylene or copolymers of ethylene or
10 propylene with one or more C_2 - C_8 alpha-olefin monomers. The preferred alpha-olefin monomers are ethylene, propylene, butene-1, pentene-1, hexene-1, 4-methylpentene-1, and octene-1. An example of a higher olefin monomer that can be copolymerised with the
15 primary ethylene and/or propylene monomer, or as partial replacement for the C_2 - C_8 monomer is decene-1. Also dienes are suitable, like 1,4-butadiene, 1,6-hexadiene, dicyclopentadiene, ethylidene norbornene and vinyl norbornene.

20 When the process is used for the copolymerisation of ethylene and/or propylene with other alpha-olefin monomers the ethylene and/or propylene are present as the major component of the copolymer, and preferably are present in an amount at
25 least 70 wt %, more preferably 80 wt % of the total monomers.

The process is particularly suitable for polymerising olefin monomers at a pressure of between 0.5 and 10 Mpa, preferably between 1 and 5 Mpa, and
30 and at a temperature of between 30°C and 130°C, and particularly between 45°C and 110°C.

The polymerisation reaction may be carried

out in the presence of any catalyst system known in the art (for instance, anionic catalyst, cationic catalyst or free-radical catalyst) suited for the gas phase polymerisation of one or more (olefin) monomers, like a catalyst system of the Ziegler-Natta type, consisting of a solid catalyst essentially comprising a compound of a transition metal and of a cocatalyst comprising an organic compound of a metal (i.e. an organometallic compound, for example an alkylaluminium compound); also so-called single site catalyst systems, like metallocene based catalyst systems, are suitable.

The catalyst may also be in the form of a prepolymer powder prepared in a prepolymerisation stage with the aid of a catalyst system described above. The prepolymerisation may be carried out by any known process, for example, polymerisation in a liquid hydrocarbon diluent or in the gas phase using a batch process, a semi-continuous process or a continuous process.

The invention also relates to a reactor system, suitable for carrying out the process of the present invention. Such a reaction system comprises a fluidised bed reactor, having at the underside a gas distribution plate, having means for the supply of reaction ingredients, having means for withdrawal of a gaseous stream from the top of the reactor, having a cooler/condensor for cooling said gaseous stream to a point where the stream partially condenses into a liquid, and having means for recirculating the stream out of the cooler/condensor to the reactor.

Such a reactor system is known from the art cited above.

The aim of the invention is to provide a reactor system, in which a process for the polymerisation of one or more (olefin) monomers is possible, in which system a higher condensed mode can
5 be applied.

This is achieved in a reactor system, wherein in the reactor the reaction zone is divided into two or more compartments by one or more substantially vertical partition walls, extending from
10 a point located above the gas distribution plate to a point located below the virtual end surface of the fluidised bed under polymerisation conditions.

In particular, said partition wall is a pipe or hollow section, preferably concentric with the
15 reactor. The preferred configurations of the reactor system of the invention are described in greater detail earlier in this specification. In particular, the reactor system of the present invention comprises means for recirculating the stream out of the
20 cooler/condensor to the reactor as a gas/liquid mixture. In another preferred mode, the reactor system also comprises a gas-liquid separator to separate at least part of the condensed liquid out of the resulting two-phase stream from the cooler/condensor and means
25 for introducing at least part of the separated liquid into the fluidised bed reactor.

It should also be appreciated that the present invention is suitable for retrofitting existing reactors by installing one or more partition walls,
30 pipes or hollow sections into the reactor. In particular, a reactor could be retrofitted by installing a pipe, as discussed above, by fixedly

attaching the pipe to an internal section of the reactor.

The invention is applicable both for so-called "grass-root" installations as well as for debottlenecking existing fluidised bed polymerisation installations. In the last case the full benefit of the invention might not be obtainable, as the capabilities of other units in the total polymerisation system may form a constraint on the maximum productivity of the system. (In other words: the throughput of the polymerisation system as a whole might be hindered by constraints in the system other than in the reactor section.) In situations where a new, integrated, polymerisation process is designed and built ("grass root"), the benefits of the present invention can be fully used and exploited.

The invention is elucidated by means of the following drawings, which are not intended to limit the boundaries of the invention.

Fig. 1 is an arrangement for polymerising one or more monomers, which includes a first embodiment of the process of the present invention with a reactor having a tubular partition wall and with a common inlet for gas and liquid below the gas distribution plate;

Fig. 2 is a similar arrangement which includes a second embodiment of the process of the invention in which gas and liquid are separately supplied to the reactor;

Fig. 3 is an arrangement as in Fig. 2, in which liquid is supplied through the wall of the reaction compartment and in which the tubular partition wall has a conical shape;

Fig. 4 is an arrangement as in Fig. 3 with a vertical plate serving as partition wall and in which liquid is introduced through the reactor wall at different heights;

5 Fig. 5 is a radial cross section of the reactor in Fig. 4 along line A-A; and

Fig. 6 is a similar cross section of a reactor in which a folded vertical plate serves as partition wall.

Fig. 1 shows a reactor body in the form of
10 a vertical cylinder in which 2 is a gas distribution plate which brings about the desired distribution of the fluidising gas introduced into the reactor below the gas distribution plate 2 through feed line 3. In the reaction zone proper 4 the gas stream introduced
15 maintains above the gas distribution plate 2 a fluidised bed of growing polymer particles which extends to below or even to the bottommost part of velocity-reducing zone 5. A cylindrical pipe 6 is concentrically suspended in reaction zone 4 from the
20 wall of reactor 1 with supports 7. Pipe 6 is submerged in the fluidised bed. Zone 5 widens relative to reaction zone 4. In this zone 5 the gas velocity decreases to the point where the gas is unable to substantially entrain any further the polymer particles
25 that have formed in the reaction zone. As a consequence, the recycle stream discharged through discharge line 8 is virtually free from entrained polymer particles. The recycle stream is cooled in heat exchanger 9, compressed in compressor 10 and cooled in
30 heat exchanger 11 to a temperature such that a proportion of the recycle stream condenses to form a two-phase stream. Make-up monomer is added to this two-

phase stream through line 12, whereupon the gas-liquid mixture is reintroduced at the bottom of the reactor through line 3. A polymer-gas stream is discharged from the reactor through drain line 13, which can be closed
5 by valve 14. This stream is separated into polymer and gaseous components in separator 15. The polymer is discharged from separator 15 through the bottom and is processed further. Such further processing may involve processes that are known per se and are not shown in
10 the figure, like removal of absorbed or dissolved liquid.

The gaseous components, on being pressurised to the required pressure, are added to the recycle stream in line 8 (not shown in the figure).
15 Besides the monomer, the required catalyst system and, optionally, an activator are supplied to the reactor. Here, too, the catalyst system is preferably supplied directly to the fluidised bed above the gas distribution plate from storage vessel 16 and propelled
20 by an inert gas through line 17, which also terminates below the lower end of pipe 6. The activator can be added to the feed stream in line 3 via metering device 18. If necessary, a propellant is also added to this stream via feed line 19.

25 The arrangement in Fig. 2 differs from the one in Fig. 1 in that the two-phase stream that evolves in the second cooling step in heat exchanger 211 is passed to gas-liquid separator 225. In this separator gas and liquid in the two-phase stream are separated
30 from each other. On adding make-up monomer and, optionally, inert gas via the feed lines 212 and 219, respectively, the separated gas stream is supplied to

the bottom of reactor 201 via feed line 203. Via line 226, which extends through gas distribution plate 202 into the fluidised bed to just below the lower end of pipe 206, the separated condensed liquid stream is
5 supplied to the fluidised bed via atomiser 227. Atomiser 227 atomises the liquid in the central reactor compartment which is located within pipe 206.

In Fig. 3, contrary to Fig. 2, the liquid separated in gas-liquid separator 325 is supplied to
10 the fluidised bed via a number of lines whose ends are symmetrically arranged along the circumference of the reactor wall at a height between the gas distribution plate 302 and the lower end of pipe 306. Two such lines, 328 and 329, are shown in the figure. The lines
15 pass through the reactor wall into reaction compartment 304 and are terminated with atomisers 330 and 331. Through these atomisers the liquid, finely atomised with the aid of fresh monomer as propellant, is introduced into the fluidised bed at such exit velocity
20 that the liquid is entrained by the fluidised bed through the central compartment located within pipe 306. Also, pipe 306 has a conical shape here with an apex angle of 1.5° . The conical shape is shown exaggerated in the purpose of clarity.

25 In Fig. 4, 440 is a vertical plate which serves as a partition wall to divide the reactor into two unequal compartments 441 and 442. A bird's eye view of this is shown in Fig. 5. In Fig. 4 a number of liquid inlets 430 pass through the reactor wall at
30 different heights to terminate in compartment 441. Catalyst feed line 417 also terminates in this compartment. A gas-impervious screen 443 extends from

gas distribution plate 402 along the inner circumference of the reactor to the reactor wall at an angle to the plate of 50°.

In Fig. 6, the partition wall plate 640 is
5 folded.

The invention is further elucidated by means of the following computer simulated Examples and comparative experiments, which are not meant to restrict the invention thereto.

10

Examples and comparative experiments

A continuous polymerisation of propylene to polypropylene is performed in a vertical, cylindrical fluidised bed reactor with an inner diameter of 0.85 m.
15 The distance between the gas distribution plate and the top of the reactor is 8.5 m.; the fluidised bed has a height of 4.2 m.

As catalyst system, a fourth generation heterogeneous Ziegler/Natta catalyst system is used,
20 the catalyst having an average particle size of 20 µm.

In all the Examples, a concentric cylindrical pipe with a diameter of 0.59 m., a length of 3.2 m., and a wall thickness of 2×10^{-3} m., is placed in the fluidised bed, at 0.4 m. above the gas
25 distribution plate. This pipe is absent in the comparative experiments.

The catalyst system, propylene, hydrogen, nitrogen (and an inert coolant) are continuously fed to the reactor; the off gas of the reactor is cooled to a
30 temperature below its dewpoint and recirculated to the bottom of the reactor. A stream of polymer product is withdrawn from the bottom of the fluid bed. The

superficial gas velocity in the fluid bed is maintained at 0.6 m/s.

During the polymerisation the maximum percentage of condensed mode (MCM, in %) is determined; this is the point at which operation of the reaction, and especially the reaction temperature, becomes unstable, and problems with the polymer withdrawal are encountered. The condensed mode ratio is the ratio of the weight of the liquid versus the weight of gas and liquid in the two-phase recycle stream to the reactor. The percentage of condensed mode can be varied by changing the cooling temperature of the recycle stream.

The process conditions and the resulting MCM and reactor productivity are given in Table I. In Examples I and III and comparative experiments A and B propylene is used as the condensable agent; in Example II a mixture of propylene and isobutane (IB) is used as the condensable agent; in Example IV a mixture of propylene and isopentane (IP) is used as the condensable agent. The composition of the gasphase is in-line monitored by means of gas chromatography.

Table I

Example/ comp. exp.	Total pressure (MPa)	C ₃ ⁺ pressure (MPa)	H ₂ - pressure (MPa)	N ₂ - pressure (MPa)	IB/IP Pressure (MPa)	Temp. reactor (°C)	MCM (%)	Temp. recycle (°C)	Produc- tivity (t/h)
I	2.50	2.16	0.04	0.30	-	70	62	37	4.8
II	2.50	2.16	0.04	0.20	0.10	70	60	48	4.7
III	2.30	1.99	0.04	0.28	-	70	61	35	4.6
IV	2.30	1.99	0.04	0.07	0.21	70	58	44	4.6
A	2.50	2.16	0.04	0.30	-	70	22	47	1.5
B	2.30	1.99	0.04	0.28	-	70	21	44	1.4

From the data it can be seen that using a process of the present invention, and more specific using the reactor system of the present invention, results in a sharp increase in the applicable condensed
5 mode, as a result of which a much higher reactor productivity is achieved.

CLAIMS

1. Process for the polymerisation of one or more
5 monomers in a fluidised bed reactor, which reactor
comprises a reaction zone which is confined at the
underside by a gas distribution plate and at the
top side by a virtual end surface, in which a
fluidised bed is maintained between the underside
10 and the top side, and in which at least part of
the gaseous stream withdrawn from the top of the
reactor is cooled to a point where the stream
partially condenses into a liquid and in which at
least part of the resulting two-phase stream is
15 recycled to the reactor via an inlet which
terminates in the reactor below the gas
distribution plate, characterised in that the
reaction zone of the reactor is divided into two
or more compartments by one or more substantially
20 vertical partition walls extending from a point
located above the gas distribution plate to a
point located below the end surface.
2. Process according to Claim 1, characterised in
that the partition wall has the shape of a pipe or
25 hollow section.
3. Process according to Claim 2, characterised in
that the pipe or hollow section is concentric with
the reaction zone.
4. Process according to Claim 1, characterised in
30 that the partition wall is a substantially axially
oriented flat, curved or folded plate.
5. Process according to any one of Claims 1-4,

characterised in that the resulting two-phase stream is recycled to the reactor as a gas-liquid mixture.

6. Process according to Claim 1, characterised in
5 that at least part of the condensed liquid is separated from the two-phase stream and directly introduced into the fluid bed.
7. Process according to any one of Claims 1-6,
characterised in that the H/D-ratio of the fluid
10 bed is greater than 5.0.
8. Process according to anyone of Claims 5-6,
characterised in that the mass ratio of (liquid
supplied to the reactor) : (the amount of gas
supplied to the reactor) is higher than 2:1.
- 15 9. Process according to anyone of Claims 1-8,
characterised in that at least one of the monomers
is ethylene or propylene.
10. Process according to anyone of Claims 1-9,
characterised in that the polymerisation is
20 performed at a pressure of between 0.5 and 10 MPa.
11. Process according to anyone of Claims 1-10,
characterised in that the polymerisation is
performed at a temperature of between 30 and 130°C.
12. Reactor system, suitable for polymerising one or
25 more monomers, comprising a fluid bed reactor,
having at the underside a gas distribution plate,
having means for the supply of reaction
ingredients, having means for the withdrawal of a
gaseous stream from the top of the reactor, having
30 a cooler/condensor for cooling said gaseous stream
to a point where said stream partially condenses
into a liquid, and having means for recirculating

- the stream out of the cooler/condensor to the reactor, characterised in that in the reactor the reaction zone is divided into two or more compartments by one or more substantially vertical partition walls, extending from a point above the gas distribution plate.
- 5
13. Reactor system according to Claim 12, characterised in that the partition wall is a pipe or hollow section.
- 10
14. Reactor system according to Claim 13, characterised in that the pipe or hollow section is concentric with the reactor.
15. Reactor system according to anyone of Claims 12-14, characterised in that the H/D-ratio of the reactor is greater than 5.
- 15
16. Reactor system according to anyone of Claims 12-15, characterised in that the ratio of the area of the radial cross section of the pipe or hollow section to that of the reactor is between 1:5 and 3:4.
- 20
17. Reactor system according to Claim 12, characterised in that the partition wall is a substantially axially oriented flat, curved or folded plate.
- 25
18. Reactor system according to anyone of Claims 12-17, characterised in that the reactor system comprises means for recirculating the stream out of the cooler/condensor to the reactor as a gas-liquid mixture.
- 30
19. Reactor system according to anyone of Claims 12-17, characterised in that the reactor system comprises a gas-liquid separator to separate at

least part of the condensed liquid out of the
resulting two-phase stream from the
cooler/condensor and means for introducing at
least part of the separated liquid into the
5 fluidised bed reactor.

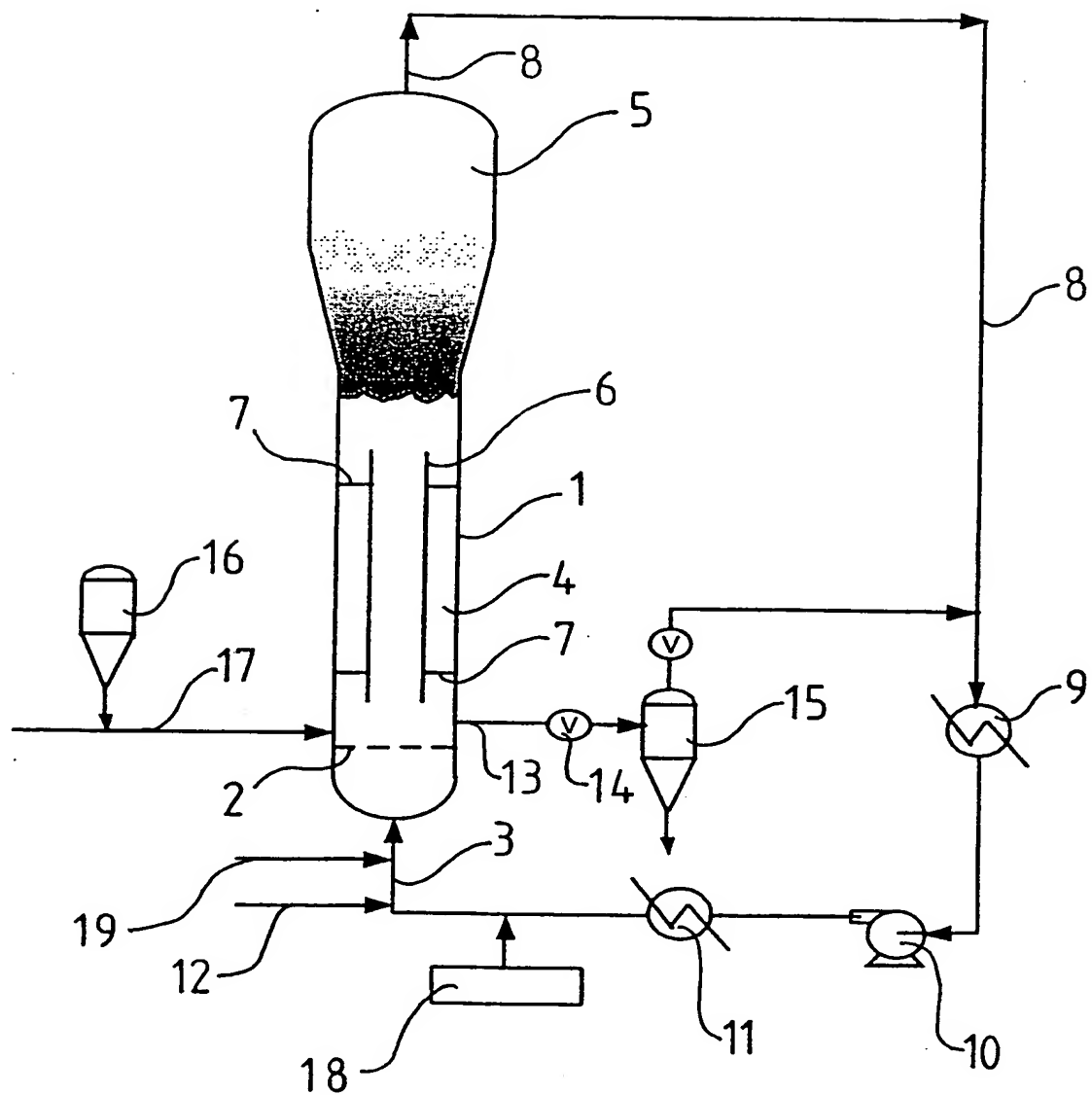


Fig. 1

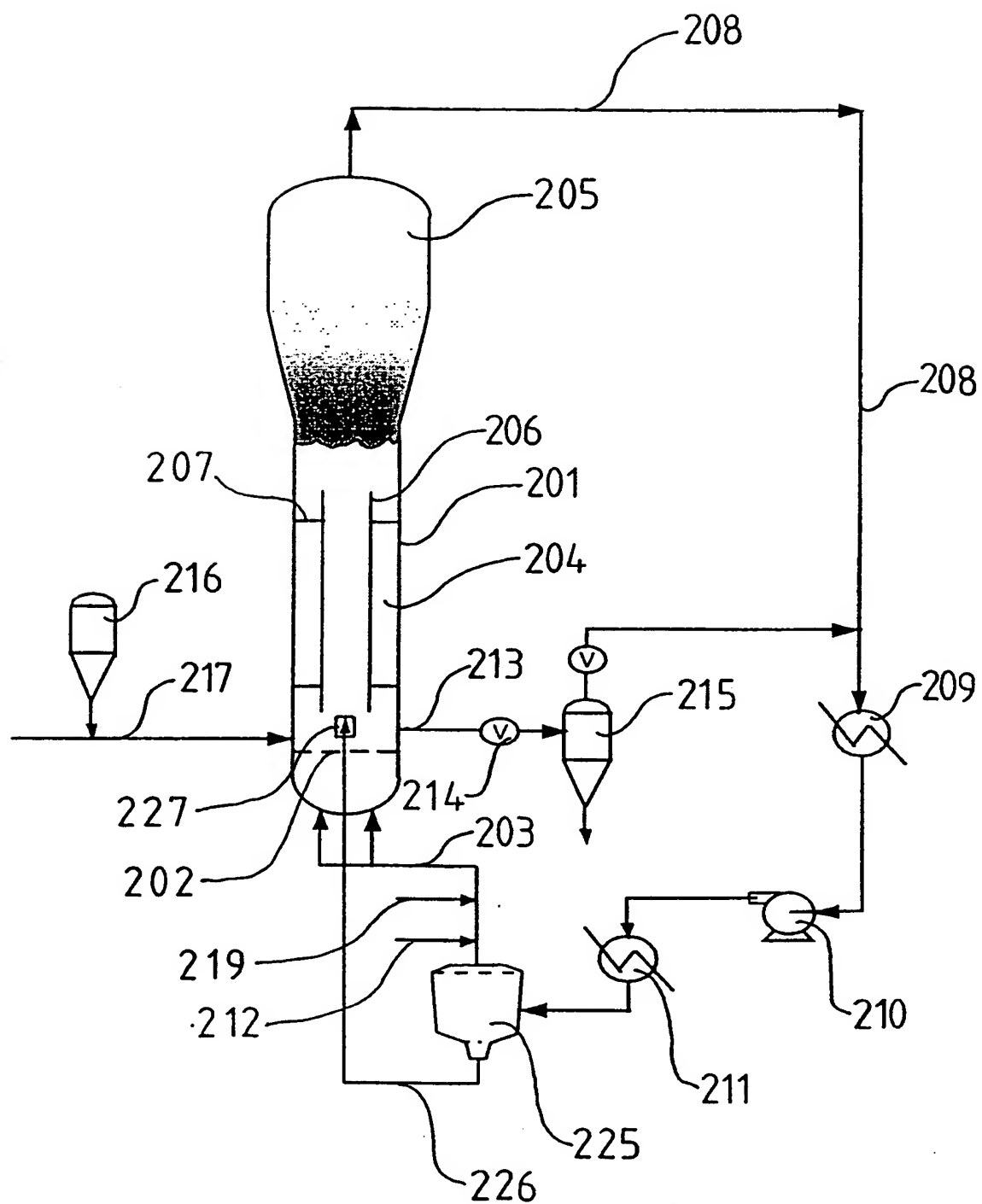


Fig. 2

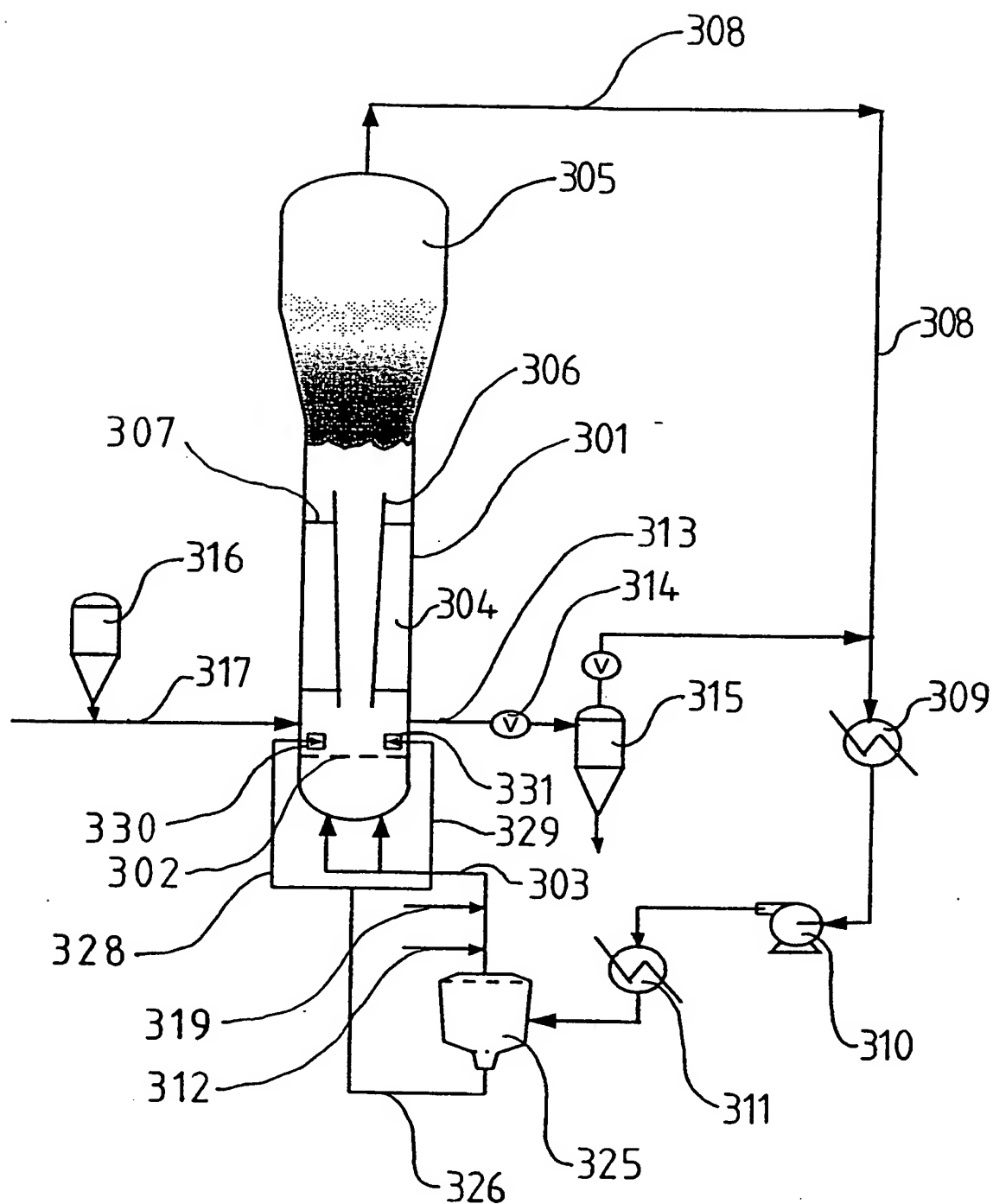


Fig. 3.

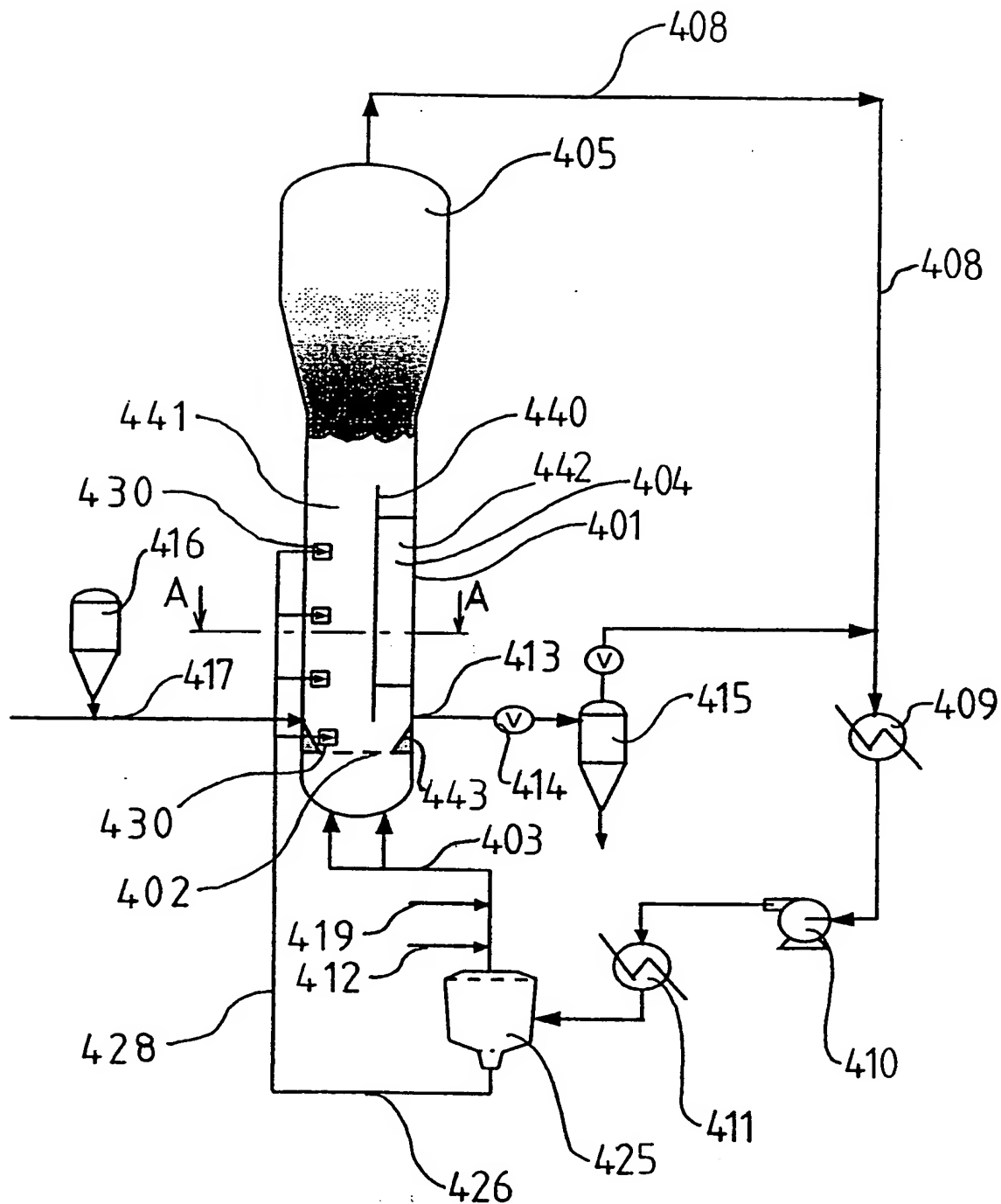


Fig. 4

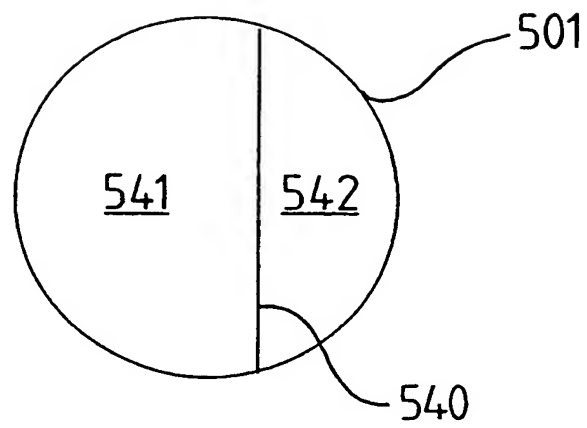


Fig. 5

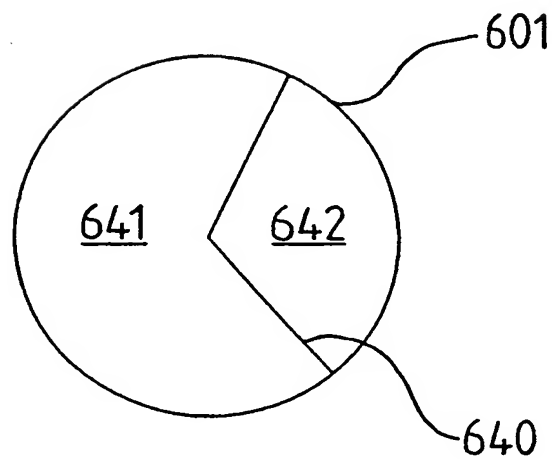


Fig. 6

INTERNATIONAL SEARCH REPORT

International Application No
PCT/NL 00/00334

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B01J8/24 B01J8/34 C08F10/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B01J C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 114 700 A (MEIHACK WOLFGANG F A T ET AL) 19 May 1992 (1992-05-19) column 4, line 51 -column 6, line 26; figures 1-6	1-4, 12-14, 17
X	EP 0 115 019 A (BASF AG) 8 August 1984 (1984-08-08) figures 1-2B	1-3, 12-14
X	GB 820 447 A (DORR-OLIVER) 23 November 1959 (1959-11-23) the whole document	1, 4
X	US 2 906 696 A (P. W. GARBO ET AL) 29 September 1959 (1959-09-29) figure 2	1-3, 12-14
	-/--	

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☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

14 August 2000

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 94 28032 A (CHINH JEAN CLAUDE ;FILIPPELLI MICHEL CHARLES HENR (FR); POWER MICH) 8 December 1994 (1994-12-08) cited in the application figures -----	1,9-12
A	GB 2 254 574 A (TAMPELLA POWER OY) 14 October 1992 (1992-10-14) figure 1 -----	1,12

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/NL 00/00334

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5114700 A	19-05-1992	AU 581103 B AU 6299886 A CA 1277822 A ZA 8607101 A	09-02-1989 26-03-1987 18-12-1990 27-05-1987
EP 0115019 A	08-08-1984	DE 3248502 A AT 26930 T DE 3371294 D JP 59173129 A US 4576573 A	05-07-1984 15-05-1987 11-06-1987 01-10-1984 18-03-1986
GB 820447 A		NONE	
US 2906696 A	29-09-1959	NONE	
WO 9428032 A	08-12-1994	AT 163017 T AT 186056 T AU 694924 B AU 6726094 A AU 701999 B AU 8077298 A BG 100102 A BR 9406535 A CA 2161432 A CN 1124029 A CZ 9502940 A DE 69408450 D DE 69408450 T DE 69421418 D DE 69421418 T EG 20361 A EP 0699213 A EP 0802202 A EP 0926163 A ES 2113104 T ES 2138430 T FI 955561 A GR 3025973 T GR 3031385 T HK 1008963 A HU 73870 A,B JP 8510497 T NO 954648 A NZ 266173 A PL 311280 A SG 49037 A SK 143395 A US 5541270 A US 5733510 A US 5668228 A US 5804677 A US 6001938 A ZA 9403399 A	15-02-1998 15-11-1999 06-08-1998 20-12-1994 11-02-1999 15-10-1998 29-11-1996 02-01-1996 08-12-1994 05-06-1996 14-02-1996 12-03-1998 20-05-1998 02-12-1999 10-02-2000 31-01-1999 06-03-1996 22-10-1997 30-06-1999 16-04-1998 01-01-2000 17-11-1995 30-04-1998 31-01-2000 21-05-1999 28-10-1996 05-11-1996 17-11-1995 25-06-1996 05-02-1996 18-05-1998 08-01-1997 30-07-1996 31-03-1998 16-09-1997 08-09-1998 14-12-1999 17-11-1995
GB 2254574 A	14-10-1992	FI 911734 A AT 402551 B AT 76292 A CN 1065716 A,B	12-10-1992 25-06-1997 15-10-1996 28-10-1992

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/NL 00/00334

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
GB 2254574 A		CS 9201047 A	14-10-1992
		DE 4212177 A	15-10-1992
		ES 2070664 A	01-06-1995
		FR 2675241 A	16-10-1992
		HU 213433 B	30-06-1997
		IT 1253997 B	05-09-1995
		PL 170574 B	31-01-1997
		SE 9200938 A	12-10-1992
		RU 2068150 C	20-10-1996
		US 5269262 A	14-12-1993

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